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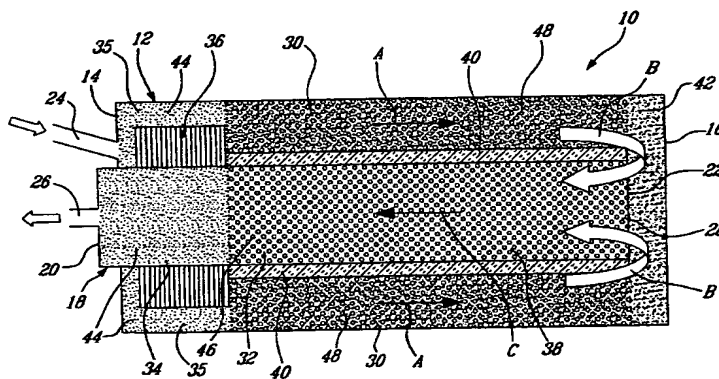
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(54) Title: **FIXED BED REACTOR FOR GAS INVOLVING EXOTHERMIC CATALYTIC REACTIONS AND PROCESS THEREFOR**



(57) Abstract: Fixed bed reactor (10) for exothermic catalytic reactions comprising a longitudinal outer chamber (12) having a proximate and a distal end (14, 16), provided with a reactor inlet (24) near the first longitudinal end (14). The reactor further comprises a longitudinal inner chamber (18) coaxially mounted in the outer chamber (12), having a proximate end and a distal end (20, 22) said inner chamber, provided with a reactor outlet (26) at its proximate end (20). The inner chamber (18) includes a) a first section (34) located near the proximate longitudinal end and being thermally coupled to the outer chamber, b) a second section (38) located near the distal longitudinal end which is in fluid communication with both the outer chamber and the first section, and c) a third section (32) located between the first and second sections which is thermally isolated (40) from the outer chamber (12). During operation, the gas entering the outer chamber (12) through the reactor inlet is preheated to the ignition temperature by the heat coming from the first section (34) of the inner chamber (18), and is forced to flow in the outer chamber in a first direction from the proximate end to the distal end. The gas then flows in the inner chamber from its distal end to its proximate end thereof, exiting through the reactor outlet.

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— Before the expiration of the time limit for amending the claims and to be republished in the event of receipt of amendments.

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

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**TITLE OF THE INVENTION**

FIXED BED REACTOR FOR GAS INVOLVING  
EXOTHERMIC CATALYTIC REACTIONS AND PROCESS THEREFOR

5

**FIELD OF THE INVENTION**

The present invention relates to catalytic gas involving  
exothermic reactions. More specifically, the present invention is concerned  
10 with a fixed bed reactor for such reactions.

**BACKGROUND OF THE INVENTION**

Traditionally, heat is produced by flame combustion of  
15 natural gas or other fossil fuels. However, this process generates nitrogen  
oxides that have noxious effects on the environment.

An environmentally preferable alternative to flame  
combustion of combustible gases or vapours is their catalytic combustion  
20 in a fixed bed reactor. However, a drawback of such method is that the  
operation of catalytic reactions involving total oxidation in classical fixed  
bed reactors becomes very difficult to control, since the reaction front  
tends to creep along the reactor axis when the concentration of fuel, as  
well as the temperature of the feed mixture, is variable.

25

Another drawback is the formation of hot spots in the  
fixed bed, leading to thermal deactivation of the catalyst.



A solution to the creep of the reaction front along the reactor axis in the case of gas streams containing relatively low concentrations of fuel has been proposed by E. J. Houdry in United States  
5 Patent No. 2,946,651, dated July 26, 1960 and entitled "Catalytic Treatment of Gas Streams". The proposed method concerns the use of a reverse-flow reactor comprising a catalytic bed and a bed of inert material serving as a heat recuperator.

10 A drawback of the reactor proposed by Houdry is that it is relatively complex and bulky and requires use of many valves and other components, increasing the overall costs of the reactor and its maintenance frequency.

15 A fixed bed reactor for catalytic exothermic reaction involving gases which has less components and is more compact than those described in the prior art is thus desirable.

20 **OBJECTS OF THE INVENTION**

An object of the present invention is therefore to provide an improved fixed bed reactor for gas involving catalytic reactions that are exothermic.

25

Another object of the present invention is to provide an improved process for exothermic catalytic gas involving reactions.





**SUMMARY OF THE INVENTION**

5 More specifically, in accordance with the present invention, there is provided a fixed bed reactor for gas involving catalytic reaction, the reactor comprising:

a longitudinal outer chamber having a proximate longitudinal end and a distal longitudinal end; the outer chamber including  
10 a reactor inlet near the proximate longitudinal end; and

a longitudinal inner chamber mounted in the outer chamber and having a proximate end and a distal end; the inner chamber including a reactor outlet at the proximate longitudinal end; the inner chamber including a) a first section located near the proximate longitudinal  
15 end and being thermally coupled to the outer chamber, b) a second section located near the distal longitudinal end and being in fluid communication with both the outer chamber and the first section, and c) a third section located between the first and second sections and being thermally insulated from the outer chamber;

20 whereby, in operation, when gas enters the outer chamber through the inlet, the gas is heated to the ignition temperature of the gas by the heat coming from the first section of the inner chamber, and is forced to flow in the outer chamber in a first direction from the proximate end to the distal end; the gas then flows in the inner chamber from the  
25 distal end to the proximate end thereof, exiting through the outlet.



Other objects, advantages and features of the present invention will become more apparent upon reading the following non-restrictive description of preferred embodiments thereof, given by way of example only with reference to the accompanying drawings.

5

### **BRIEF DESCRIPTION OF THE DRAWINGS**

In the appended drawings:

10                      Figure 1 is a schematic sectional view of a fixed bed reactor according to a first embodiment of the present invention;

15                      Figure 2 is a chart and a schematic diagram illustrating the temperature profile and the position of the reaction front in the reactor of Figure 1 for combustion of 0.88 % propane in air, total flow rate 16.95L/min (air flow rate: 16.8 L/min, propane flow rate: 0.15L/min, propane concentration: 0.88%);

20                      Figure 3 is a chart and a schematic diagram illustrating the temperature profile and the position of the reaction front in the reactor of Figure 1 for combustion of 0.67% of propane in air, total flow rate: 22.25L/min (air flow rate: 22.1L/min, propane flow rate: 0.15L/min);

25                      Figure 4 is a chart and a schematic diagram illustrating the temperature profile and the position of the reaction front in the reactor of Figure 1 for combustion of 0.55% propane in air, total flow rate: 27.55L/min (air flow rate: 27.4L/min, propane flow rate: 0.15L/min);



Figure 5 is a chart and a schematic diagram illustrating the temperature profile and the position of the reaction front in the reactor of Figure 1 for combustion of 0.46% propane in air (total flow rate: 32.95L/min, air flow rate: 32.7L/min, propane flow rate: 0.15L/min);

Figure 6 is a chart and a schematic diagram illustrating the temperature profile, the position of the reaction front and the re-ignition at the inlet of the reactor of Figure 1 for propane combustion (total flow rate: 32.85L/min., air flow rate: 32.7L/min., propane flow rate: 0.15L/min);

Figure 7 is a chart representing the time evolution of temperature profiles in the reactor of Figure 1 at the position of the thermocouples T20 and T25 for combustion of very lean propane ( $0.33 \pm 0.13\%$ ) mixture at a flow rate of 32.85L/min;

Figure 8 is a chart and a schematic diagram illustrating the temperature profile and the position of the reaction front in the reactor of Figure 1 for combustion of 5% methane in air, total flow rate 20.52 L/min (air flowrate: 19.5L/min, methane flowrate: 1.02 L/min);

Figure 9 is a chart and a schematic diagram illustrating the temperature profile and the position of the reaction front in the reactor of Figure 1 for combustion of 1.98% methane in air, total flow rate 25.2 L/min (air flowrate: 24.7L/min, methane flowrate: 0.5 L/min);



Figure 10 is a chart and a schematic diagram illustrating the temperature profile and the position of the reaction front in the reactor of Figure 1 for combustion of 1.64% methane in air, total flowrate 30.5 L/min. (air flow rate: 30 L/min, methane flow rate: 0.5 L/min);

5

Figure 11 is a chart and a schematic diagram illustrating the temperature profile, the position of the reaction front and the re-ignition at the inlet of the reactor of Figure 1 for combustion of  $1.25 \pm 0.4\%$  methane in air, total flow rate 30.5L/min;

10

Figure 12 is a chart representing the time evolution of temperature profiles at the position of the thermocouples T20 and T25 for combustion of lean methane mixture ( $1.25 \pm 0.4\%$ ) in air at a flow rate of 30.5L/min; and

15

Figure 13 is a schematic sectional view of a fixed bed reactor according to a second embodiment of the present invention

#### **DESCRIPTION OF THE PREFERRED EMBODIMENT**

20

Turning to Figure 1 of the appended drawing, a fixed bed reactor 10, according to an embodiment of the present invention, will now be described.

25

The reactor 10 comprises a generally cylindrical outer chamber 12 having opposite proximate and distal longitudinal ends 14 and 16, and a generally cylindrical inner chamber 18 having proximate and





distal longitudinal ends 20 and 22, and being coaxially provided in the outer chamber 12.

Although the outer and inner chambers 12 and 18 are  
5 described as being advantageously cylindrical and coaxial, these chambers 12 and 18 may have other configurations and respective positions without departing from the spirit of the present invention.

The outer chamber 12 is provided with a reactor inlet 24  
10 at its proximate end 14, while the inner chamber 18 is provided with a reactor outlet 26 at its distal end 22.

It is to be noted that, when the reactor 10 is used to heat  
air, the outlet 26 is advantageously connected to an air interchanger (not  
15 shown).

The distal end 22 of the inner chamber 18 includes an  
aperture 28 that provides fluid communication between the outer and inner  
chamber 12 and 18.

20

As will now be apparent to a person skilled in the art, the  
portion of the outer chamber 12 that surrounds the inner chamber 18  
defines a first passage 30 for gas entering the inlet 24 to flow in a first  
direction from the proximate end 14 to the distal end 16 of the outer  
25 chamber 12.



Similarly, the inner chamber 18 defines, from its distal end 22 to its proximate end 20, a second passage 32 for the gas to flow in a second direction generally parallel, but opposite to the direction of the flow in the first passage 30.

5

The inner chamber 18 includes a first section 34 located near its proximate end 20 and being thermally coupled with the outer chamber 12 using fins 36. Since fins are believed to be well known in the art and for concision purposes, they will not be described in more detail herein.

10

Other thermal coupling means, such as heat pipes can also be used without departing from the spirit of the present invention.

15

A second section 38, advantageously extending from the first section 34 of the inner chamber 18 to its distal end 22, is generally thermally insulated from the outer chamber 12 using an insulating layer 40 made of an insulating material such as ceramic fibers. Of course, this insulating layer 40 has the shape of the inner chamber. Other insulators can also be used.

20

To start the operation, the first section 35 of the outer chamber 12 of the reactor 10 in which the reaction mixture enters through the inlet 24 has to be preheated to the ignition temperature by auxiliary heating means, such as electrical heater (not shown).

25



It is to be noted that the ignition temperature depends on the activity of the catalyst in the fixed bed. The catalyst 44 in the first section 35 of the outer chamber 12 is advantageously very active. The heater is shut off immediately after the reaction has been initiated, since  
5 the heat for ignition of the entering gas will be further provided through the fins 36.

The ignited gas is forced to flow through the first passage 30 in the outer chamber 12 in a first direction (as shown by  
10 arrows A) from the proximate end 14 to the distal end 16.

The gas is then forced into the second section 38 of the inner chamber 34 via the aperture 28 (as shown by arrows B) where it flows in a second direction (see arrow C), opposite the first direction. The  
15 gas exits through the outlet 26.

Subsequently, as will now be apparent to a person skilled in the art, since the reaction in the reactor 10 is used for exothermic reaction, the temperature of the gas in the first section 34 of the inner  
20 chamber 18 is greater than the temperature of the gas entering the reactor 10 through the inlet 24. Since this first section 34 is thermally coupled via the fins 36 to the section of the outer chamber 12 where the gas enters the reactor 10, the gas entering the reactor 10 is heated by the heat of the gas flowing out of the reactor 10, and the reaction continues.

25

It is to be noted that the rate and the direction of the exothermic reaction depends on the volumetric heat balance.



If the reaction front, that is defined as the hottest part of the reactor bed 10, moves to the first section 34 of the inner chamber 18, the heat from section 34, communicated via fins 36 to the section 35 of the outer chamber 12, is sufficient to heat the catalytic bed to the temperature of reaction initiation, therefore initiating a new cycle.

Some or all of the sections of the reactor 10 may be advantageously filled with materials suitable for the desired exothermic catalytic reactions.

For example, the reactor 10 illustrated in Figure 1 includes sand 42 in the outer chamber 12 near its second end 16, catalytic particles 44, such as palladium or platinum supported on alumina pellets, in the first section 34 of the inner chamber 18 and in the first section of the outer chamber, catalytic pellets 46 in the second section 38, and a mixture of catalytic pellets and sand 48 in the section of the outer chamber 12 surrounding the second section 38 of the inner chamber 18. This configuration is suitable in catalytic combustion of lean fuel/air mixtures to produce hot air.

A variety of types of catalytic materials may be employed along with inert materials, depending upon the reactions that will occur in the reactor 10, its heat content and the size of the reactor 10.

25

Figures 2 to 7 represent experimental results for catalytic combustion of propane in air using the reactor bed 10 (outer chamber





consisting of stainless steel pipe, 2" (5.1 cm) nominal, 67 cm long; inner chamber consisting of stainless steel ½" (1.3 cm) nominal pipe, 67 cm long) and illustrate the movement of the reaction front as a function of propane concentration and total flow rate.

5

Figure 2 illustrates the temperature profile in the reactor for combustion of 0.88% propane in air at a total flow rate of 16.95L/min. One can see that, at the position of thermocouple T17, the temperature is about 250°C, reaches about 520°C (position of the reaction front) at the position T20 and then falls to about 200°C at the thermocouple T22 at the distal end 20 of the inner chamber 18. The temperature in the inner chamber 18 near the proximate end 20 at the thermocouple T25 rises to about 450°C. One can see that at these conditions (0.88% propane, 26.95L/min.) the reaction front did not move in two hours of operation.

15

Figures 3 to 5 show how the reaction front displaces for different conditions. As the concentration of propane decreases and the flow rate increases, the displacement becomes faster.

20

Figure 3 shows that for 0.67% propane at 22.25L/min. it took 13 hours for the reaction front to move from position at the thermocouple T20 to the thermocouple T27. For these conditions the front started to move and it took thirteen hours for the front to move from the position of the thermocouple T20 to the position of the thermocouple T27.

25

Figure 4 shows the movement of the reaction front for 0.55% propane in air at a flow rate of 27.55L/min. For these conditions, it



took only three hours and fifteen minutes for the front to move from the position of the thermocouple T20 to the position of the thermocouple T27.

Figure 6 shows the movement of the reaction front for  
5 0.46% propane in air at a flow rate of 32.85L/min. In this case it took the reaction front only about one hour to get displaced from the position of the thermocouple T20 to the position of the thermocouple T28.

Figure 7 shows the evolution with time of temperature  
10 profiles at the position the thermocouples T20 and T25 for combustion of very lean propane mixture ( $0.33 \pm 0.13\%$ ) at a flow rate of 32.85L/min.

Figures 8 to 12 represent experimental results for catalytic combustion of methane in air using the self regulating reactor 10  
15 (0.67 m long, 5.25 cm diameter), and illustrate the movement of the reaction front as a function of methane concentration and total flow rate.

Figure 8 illustrates the temperature profile in the reactor 10 for combustion of 5% methane in air at a total flow rate of 20.52L/min.  
20 One can see that for this high methane concentration and relatively low flow rate the reaction front is located at the position of thermocouple T17, the temperature increasing from about 480° to 620°C in an hour of operation and the front does not move.

25 Figure 9 shows the movement with time of temperature profiles for combustion of 1.98% methane in air at a total flow rate 25.2L/min. In this case the reaction front starts to displace passing in 5



hour 50 min. from position of the thermocouple T20 to that of the thermocouple T23.

Figure 12 shows the evolution with time of temperature profiles at the position of the thermocouples T20 and T25 for combustion of lean methane/air mixture ( $1.24 \pm 0.4\%$ ) at a total flow rate of air 32.7L/min.

As shown, the temperature profile of the reaction front for methane combustion is similar to that for propane combustion described above. Figures 8, 9 and 10 show that the temperature of methane moves further into the reactor 10 as the concentration of methane is reduced (5%, 1.98% and 1.64% respectively) and as the flow rate increases. Figure 11 shows that the high temperature at the thermocouple T25 re-ignites the methane mixture entering the reactor 10 causing an increase in temperature at the thermocouple T18. Figure 13 illustrates that, after re-ignition, the overall temperature of the second cycle of the reaction front is greater than that of the first cycle. Again, all of these experiments demonstrated that the temperature at the thermocouple T25, near the reactor outlet 26, was always greater than at the thermocouple T17 near the reaction inlet 24.

Hence, it is believed that a fixed bed reactor, according to the present invention and a process therefor, has demonstrated that it is possible to efficiently use its own generated heat to self-regulate gas-phase exothermic reactions.



Since a reactor according to an embodiment of the present invention, is self-regulating and self-cycling, it can be operated in any convenient position and orientation from horizontal to vertical.

5 Advantages of a fixed bed reactor according to embodiments of the present invention include: less parts or components and therefore more reliability, compactness and low maintenance.

10 Although, the operation of the reactor 10 and its use has been described as a heat generator, it can also be used in other catalytic gas involving exothermic reaction including:

- catalytic cleaning of gas streams (effluents) containing combustible gases or vapors;
- 15 • production of energy by the combustion of biogas;
- partial oxidation of hydrocarbons; and
- production of sulfur trioxide from relatively low concentrated sulfur dioxide containing gases.

20 More generally, a fixed bed reactor according to the present invention can be used, for example, as a heating system for industrial, commercial and residential use, catalytic cleaning of ambient air in mines and other closed spaces contaminated by emanating combustible gases and for VOC (Volatile Organic Contaminant) abatement.

25

Figure 13 of the appended drawings illustrates a second embodiment of a fixed bed reactor for gas involving catalytic reactions.





The reactor 100 is used as a heating system. Since the operation of reactor 100 and its general structure are similar to those of reactor 10 and for concision purposes, only differences between reactor 100 and reactor 10 will be discussed herein.

5

Similarly to the reactor 10, the reactor 100 is provided with an inlet 110 and an outlet 112 for alimentation and evacuation of a combustible mixture of air-lean gas.

10

The sections of the outer and inner chamber of the reactor 100 that are thermally coupled are filled with first catalytic particles 106, while the rest of the reactor bed is generally filled with second catalytic particles 108. The particles 106 and 108 are so chosen as to maximise the combustion of the mixtures and the heat exchange.

15

The outer chamber does not include any thermal insulation to allow the radiation of heat of the reactor through the outer chamber outer wall (illustrated by arrows 104 in Figure 13).

20

The reactor 100 is advantageously provided in an enclosure 102 that also host a fan to help evacuate heat from the reactor 100.

25

Although the present invention has been described hereinabove by way of preferred embodiments thereof, it can be modified, without departing from the spirit and nature of the subject invention, as defined in the appended claims.



**WHAT IS CLAIMED IS:**

1. A fixed bed reactor for gas involving catalytic reaction, said reactor comprising:

- 5                   a longitudinal outer chamber having a proximate longitudinal end and a distal longitudinal end; said outer chamber including a reactor inlet near said proximate longitudinal end; and a longitudinal inner chamber mounted in said outer chamber and having a proximate end and a distal end; said inner chamber including a reactor  
10   outlet at said proximate longitudinal end; said inner chamber including a) a first section located near said proximate longitudinal end and being thermally coupled to said outer chamber, b) a second section located near said distal longitudinal end and being in fluid communication with both said outer chamber and said first section, and c) a third section located  
15   between said first and second sections and being thermally insulated from said outer chamber;  
whereby, in operation, when gas enters said outer chamber through said inlet, said gas is heated to the ignition temperature of the gas by the heat coming from said first section of said inner chamber, and is forced to flow  
20   in said outer chamber in a first direction from said proximate end to said distal end; said gas then flows in said inner chamber from said distal end to said proximate end thereof, exiting through said outlet.

2. A reactor as recited in claim 1, wherein said reactor  
25   further comprising an auxiliary heating means and wherein the operation of said reactor is initiated by preheating the entering gas to the ignition temperature of said gas using said auxiliary heating means.



3. A reactor, as recited in claim 1, wherein at least one of said outer chamber and said inner chamber is cylindrical

5                   4. A reactor, as recited in claim 1, wherein said outer and inner chambers are generally concentric.

5. A reactor, as recited in claim 1, wherein said first section of said inner chamber is thermally coupled to said outer chamber  
10 via fins.

6. A reactor, as recited in claim 1, wherein said second section of said inner chamber includes an aperture for fluid communication with said outer chamber.

15                   7. A reactor, as recited in claim 1, wherein said third section of said inner chamber is covered with an insulating material.

8. A reactor, as recited in claim 1, wherein said first  
20 section of said inner chamber includes at least one of said catalytic particles, catalytic pellets and sand.

9. A reactor, as recited in claim 1, wherein said third  
25 section of said inner chamber includes at least one of said catalytic particles, catalytic pellets and sand.



10. A reactor, as recited in claim 1, wherein said outer chamber includes at least one of said catalytic particles, catalytic pellets and sand.

5                    11. A reactor, as recited in claim 1, wherein said first section of said inner chamber includes catalytic particles, said second section of said inner chamber includes catalytic pellets and said outer chamber includes sand and mixtures of sand and catalytic pellets.

10                   12. The use of the reactor of claim 1 to heat air.

13. The use of the reactor of claim 1 for catalytic cleaning of gas streams containing combustible gases.

15                   14. The use of the reactor of claim 1 for production of sulfur trioxide.

15. The use of the reactor of claim 1 for the production of energy by the combustion of biogas.

20                   16. The use of the reactor of claim 1 for partial oxidation of hydrocarbons.





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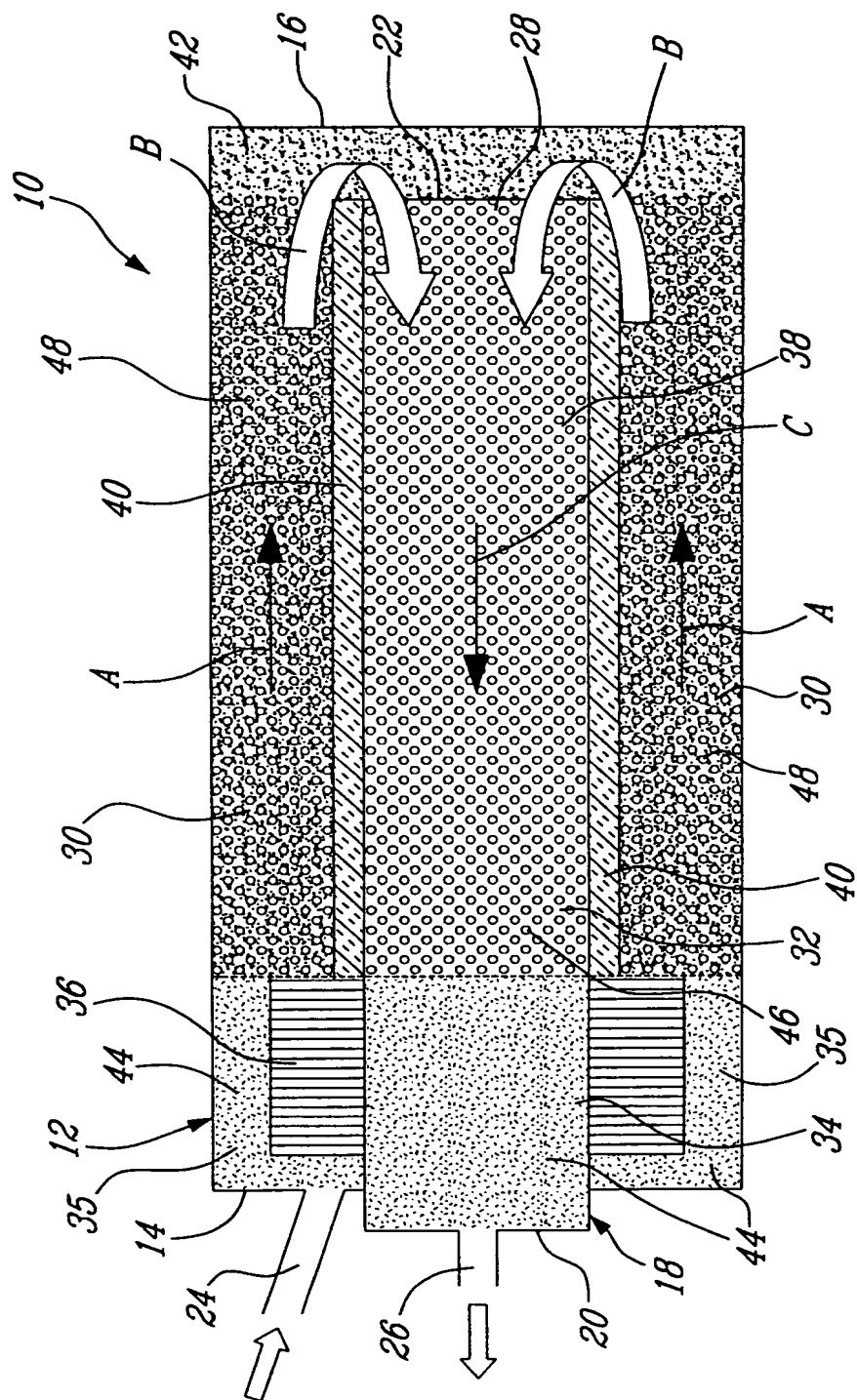
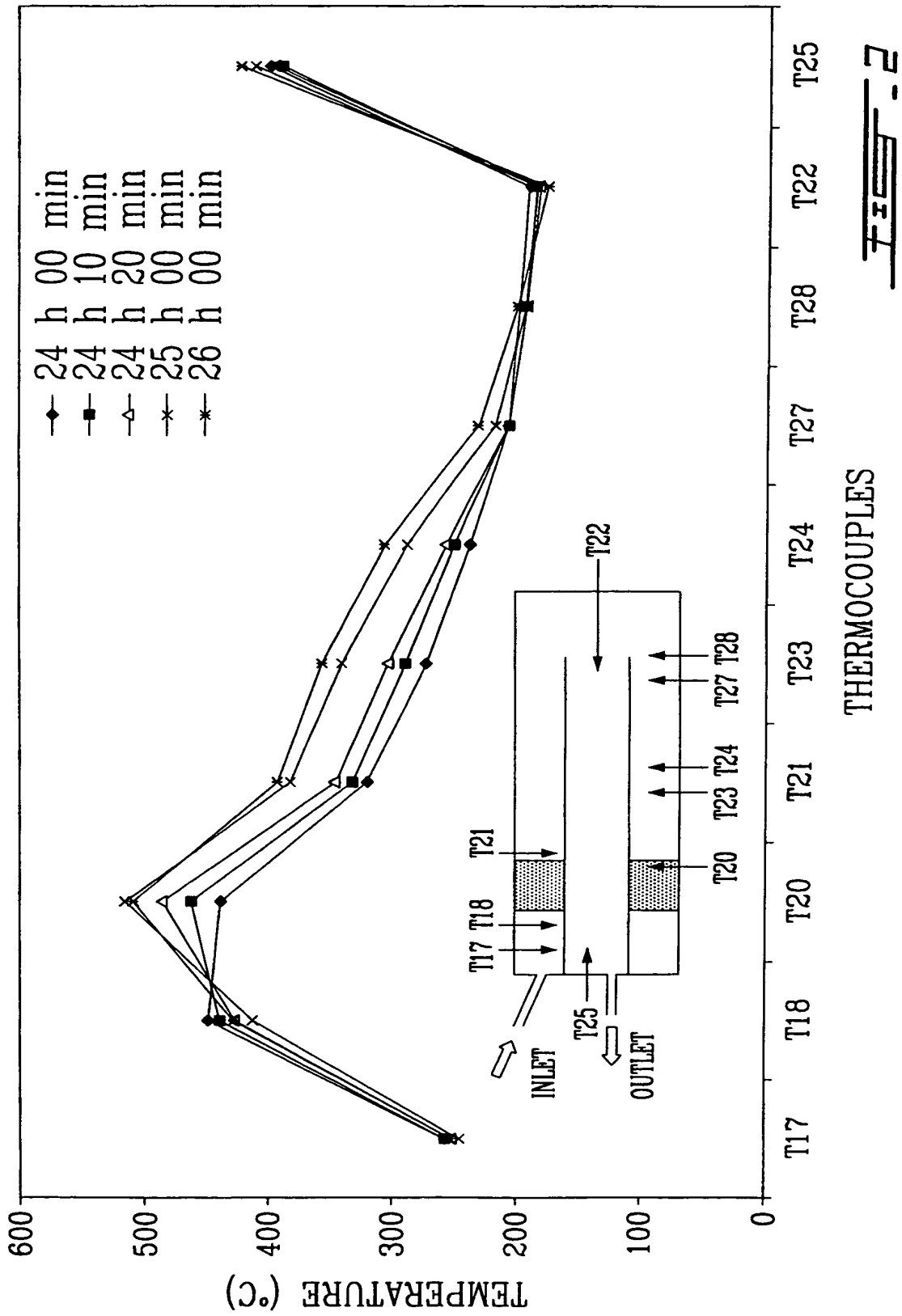


FIG. 1



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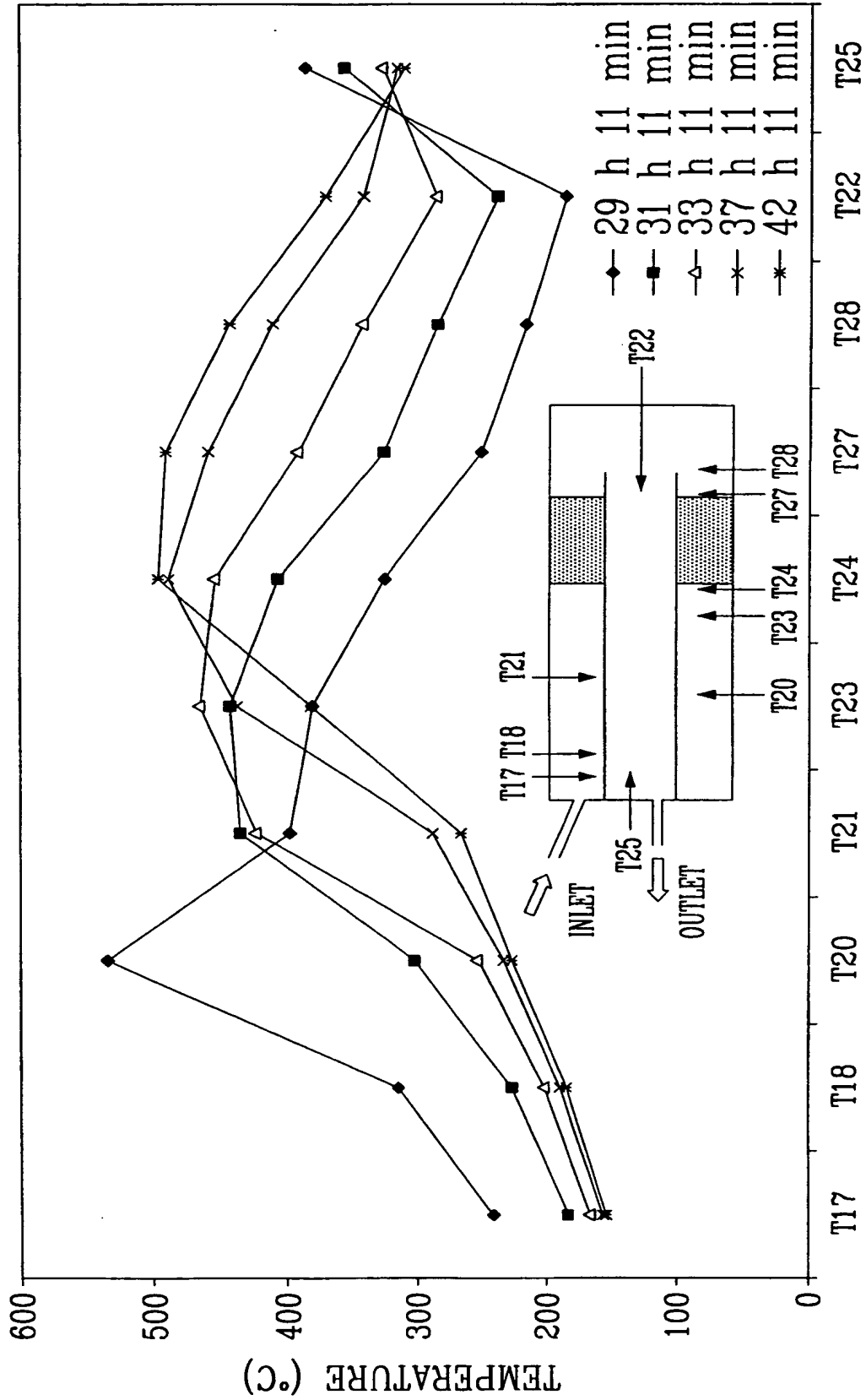


THERMOCOUPLES

Fig. 2



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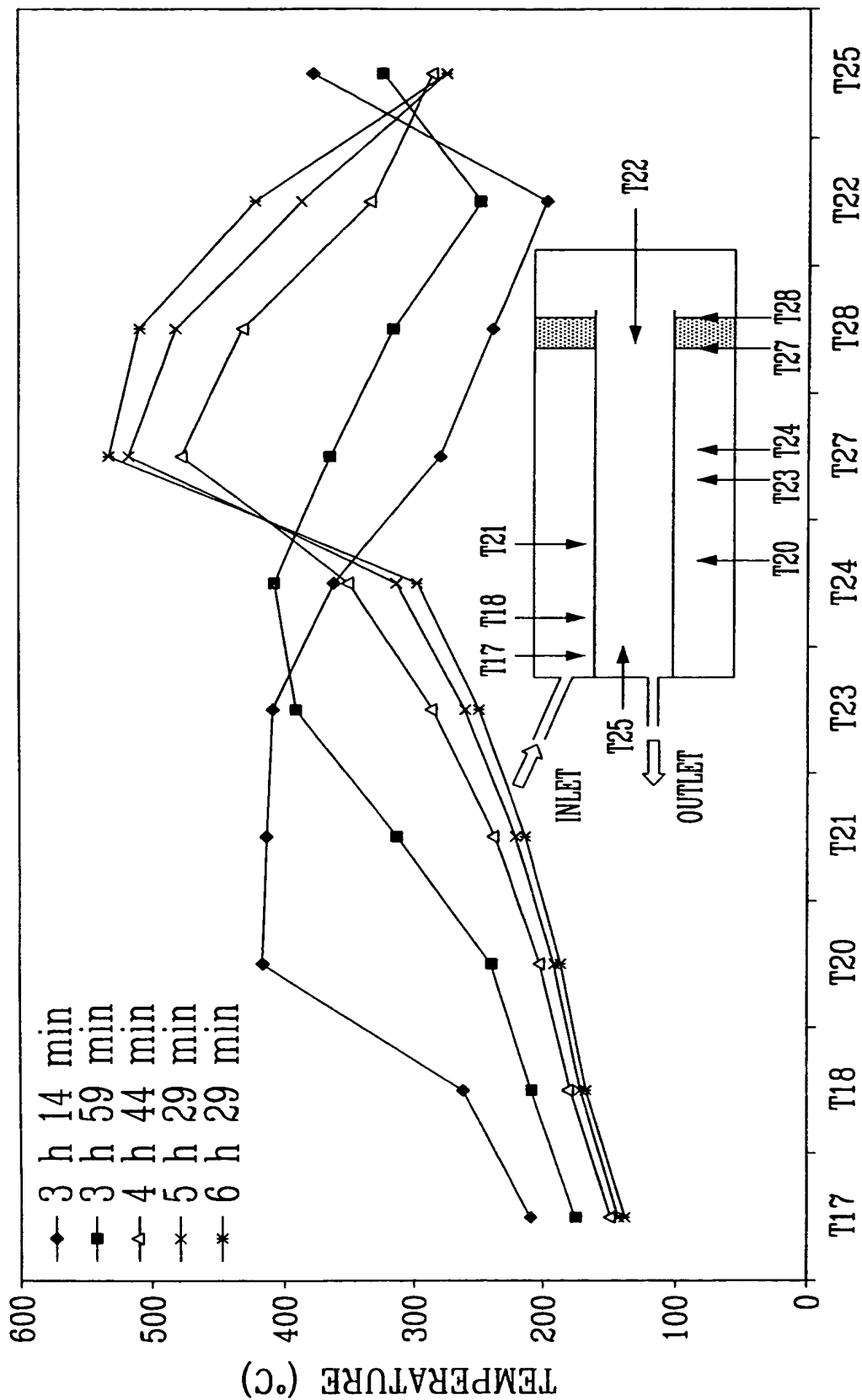


THERMOCOUPLES

Fig. 3



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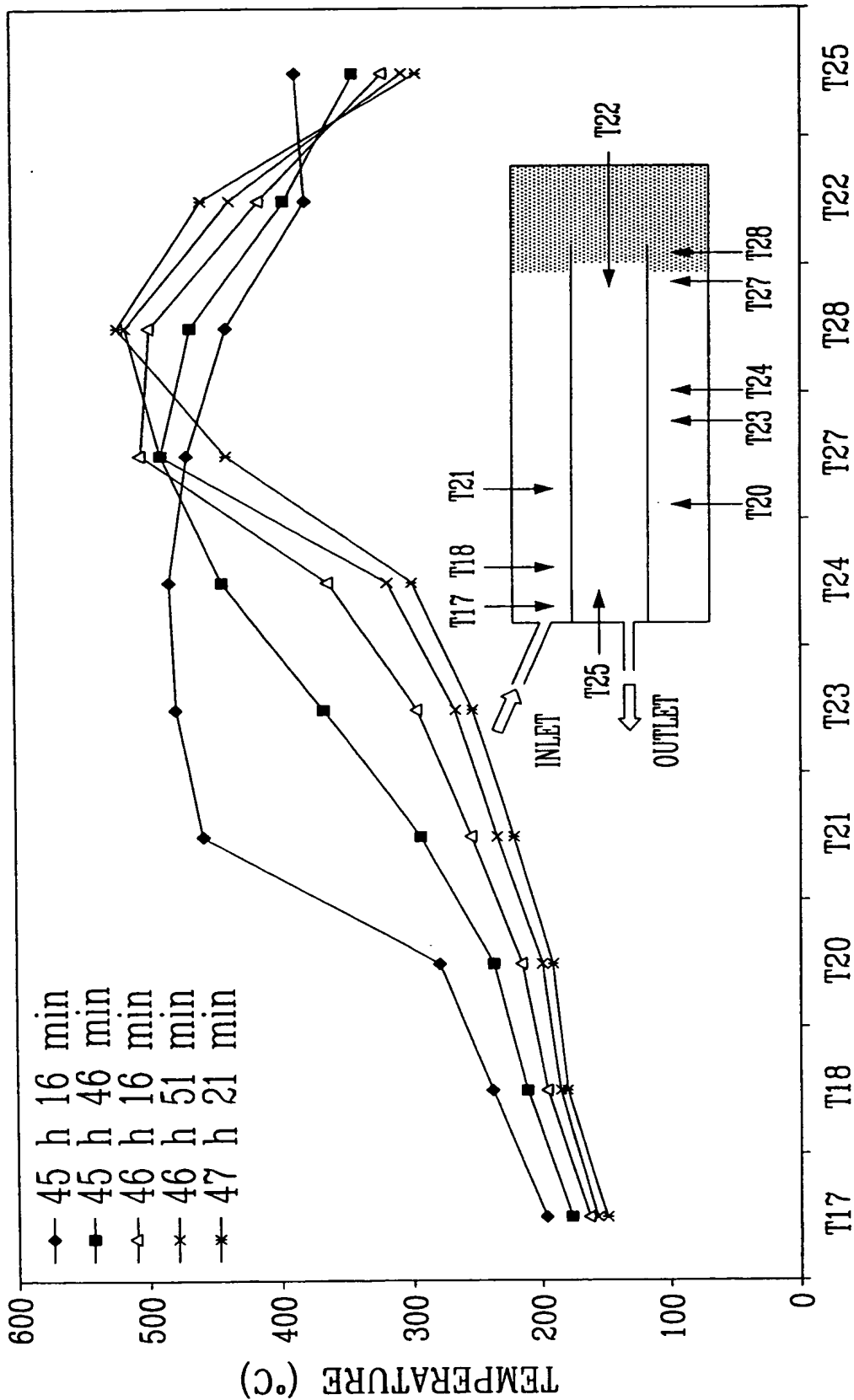
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Fig. 4





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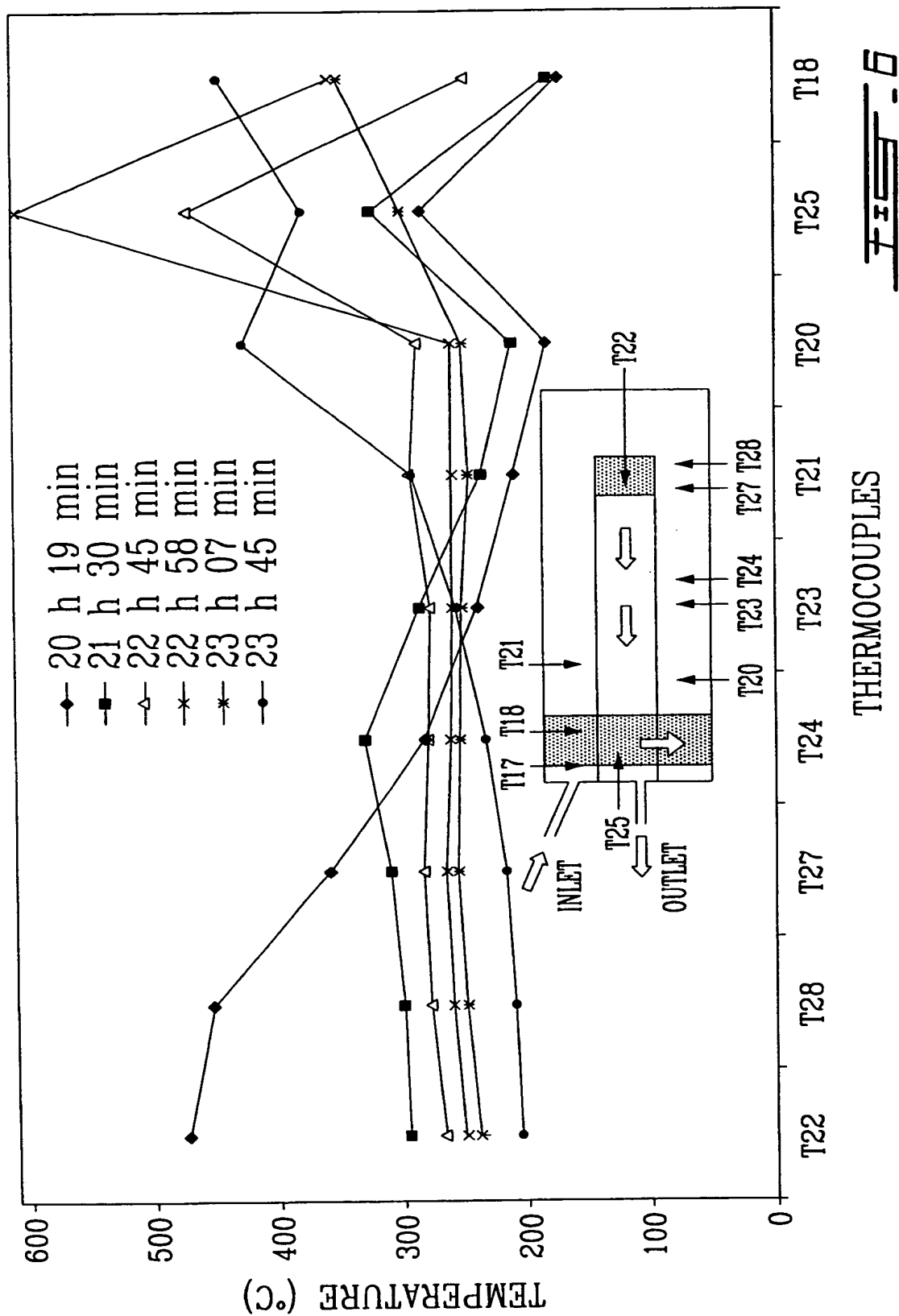


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Fig. 5

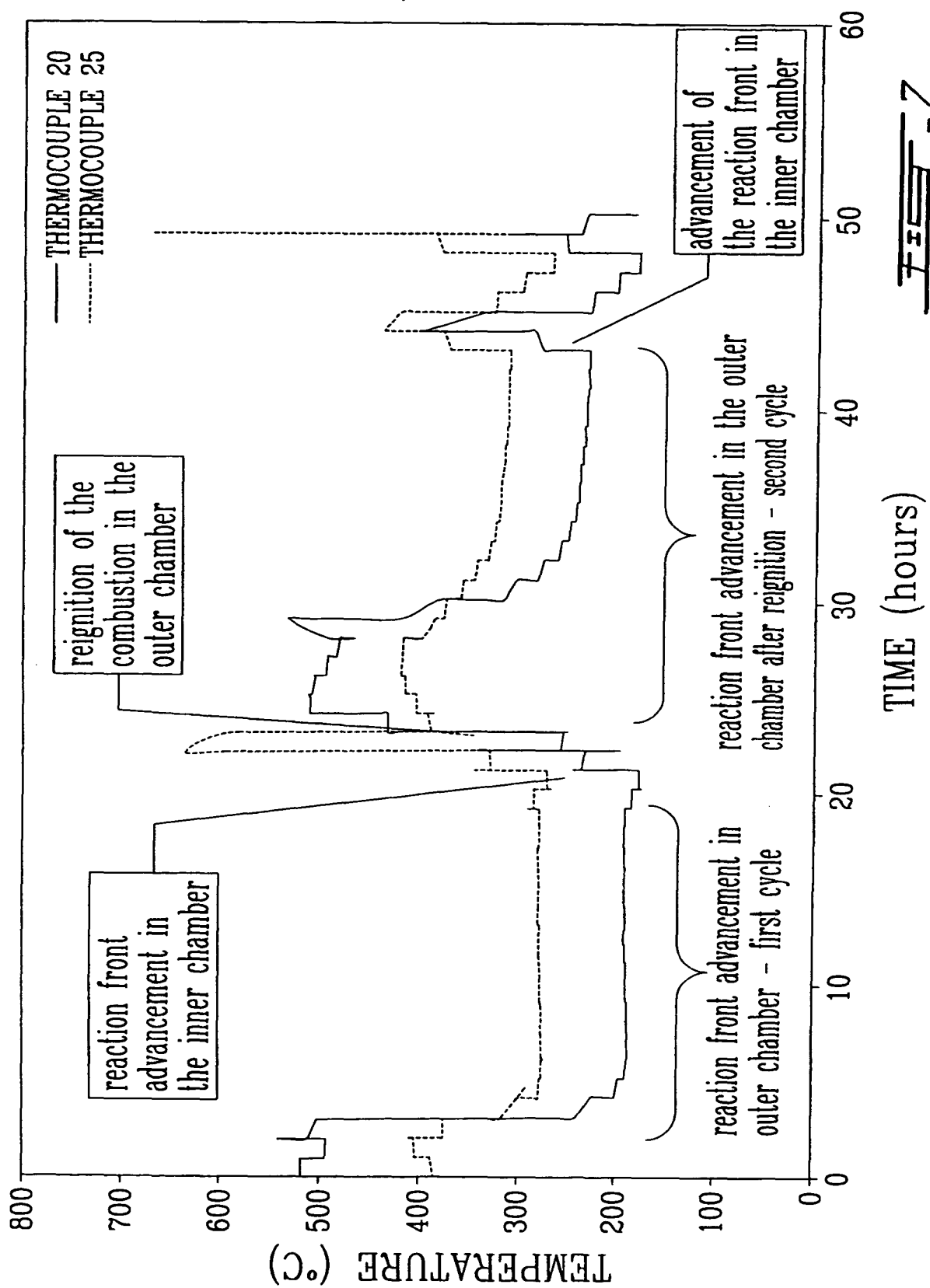


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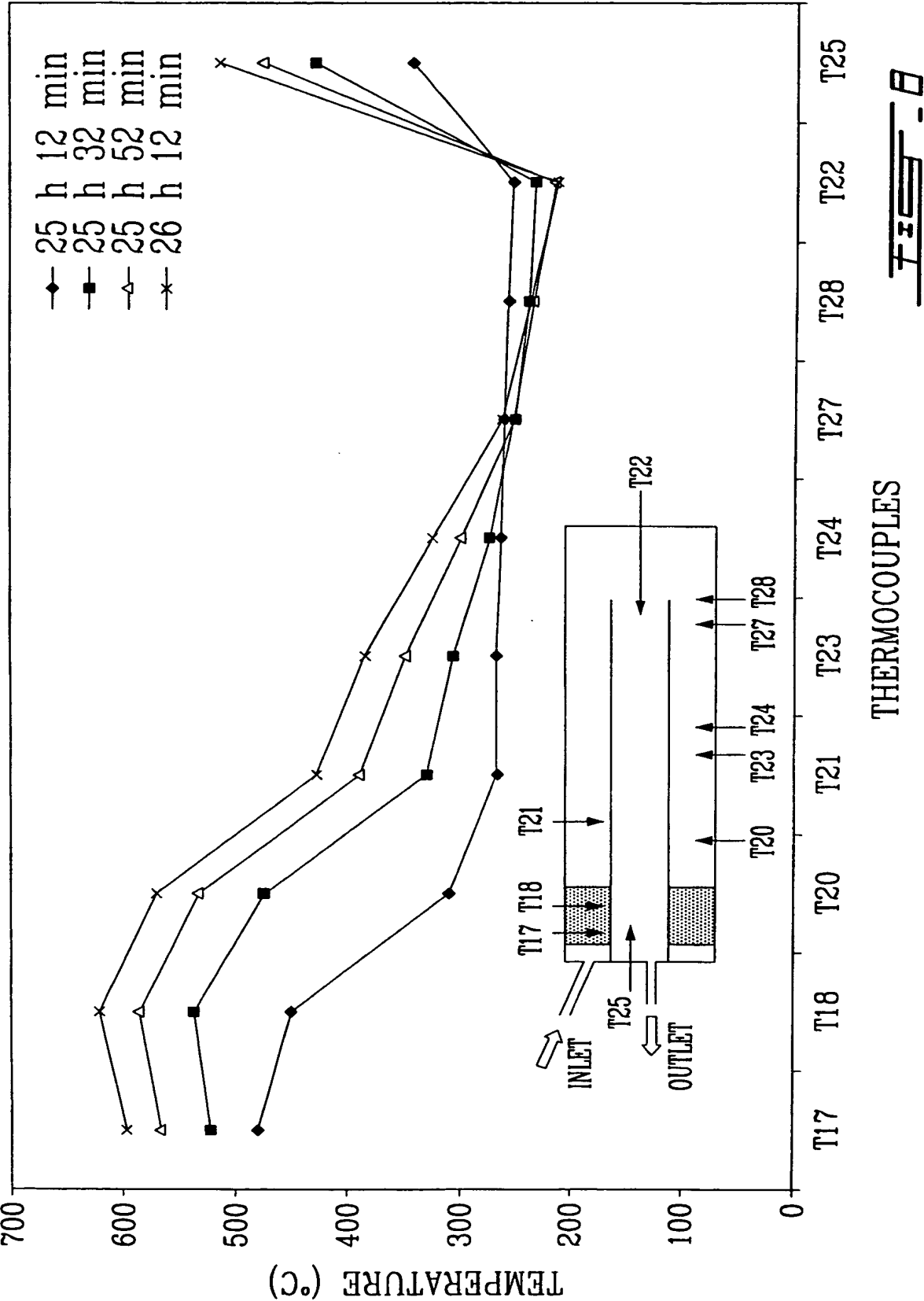


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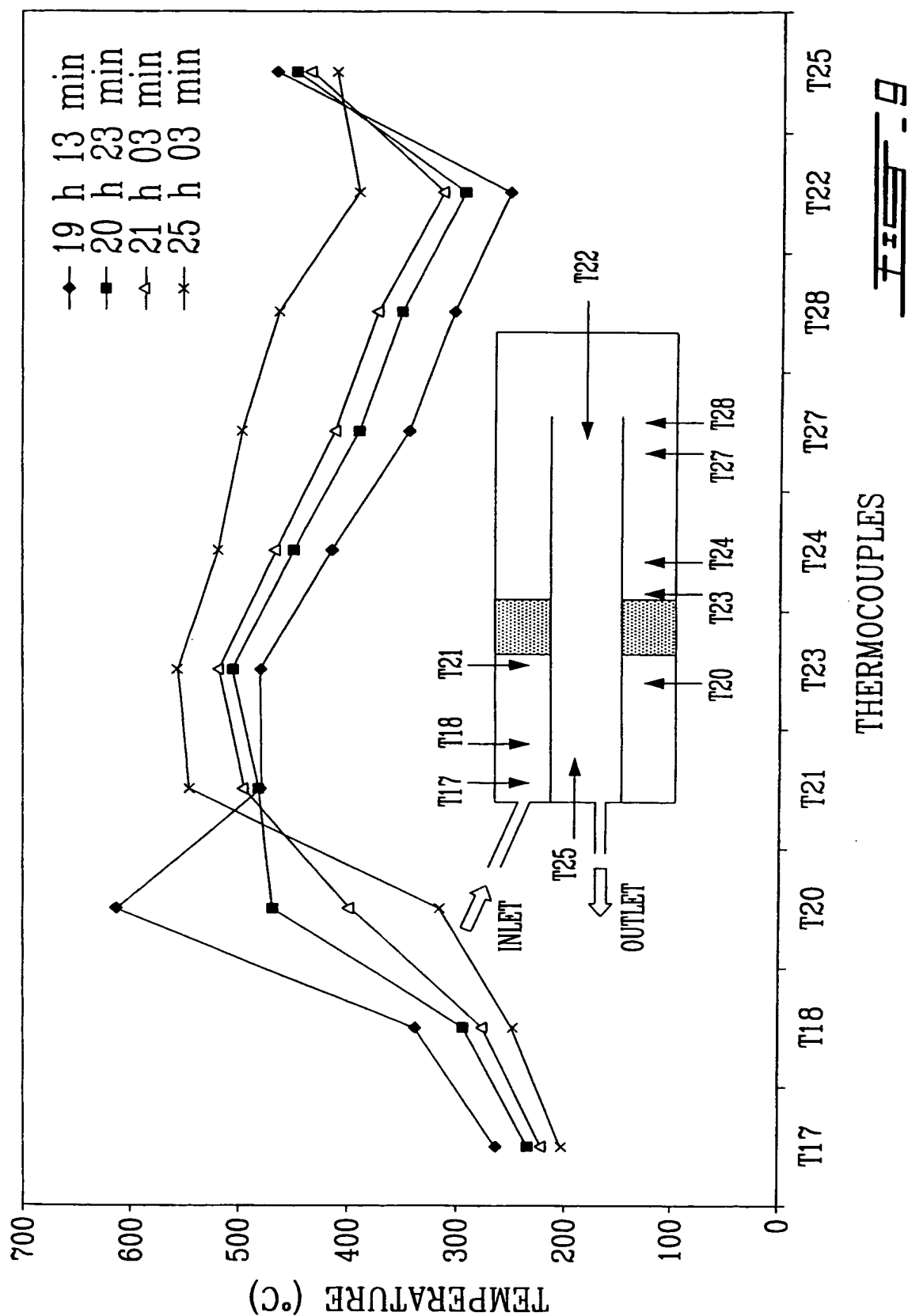
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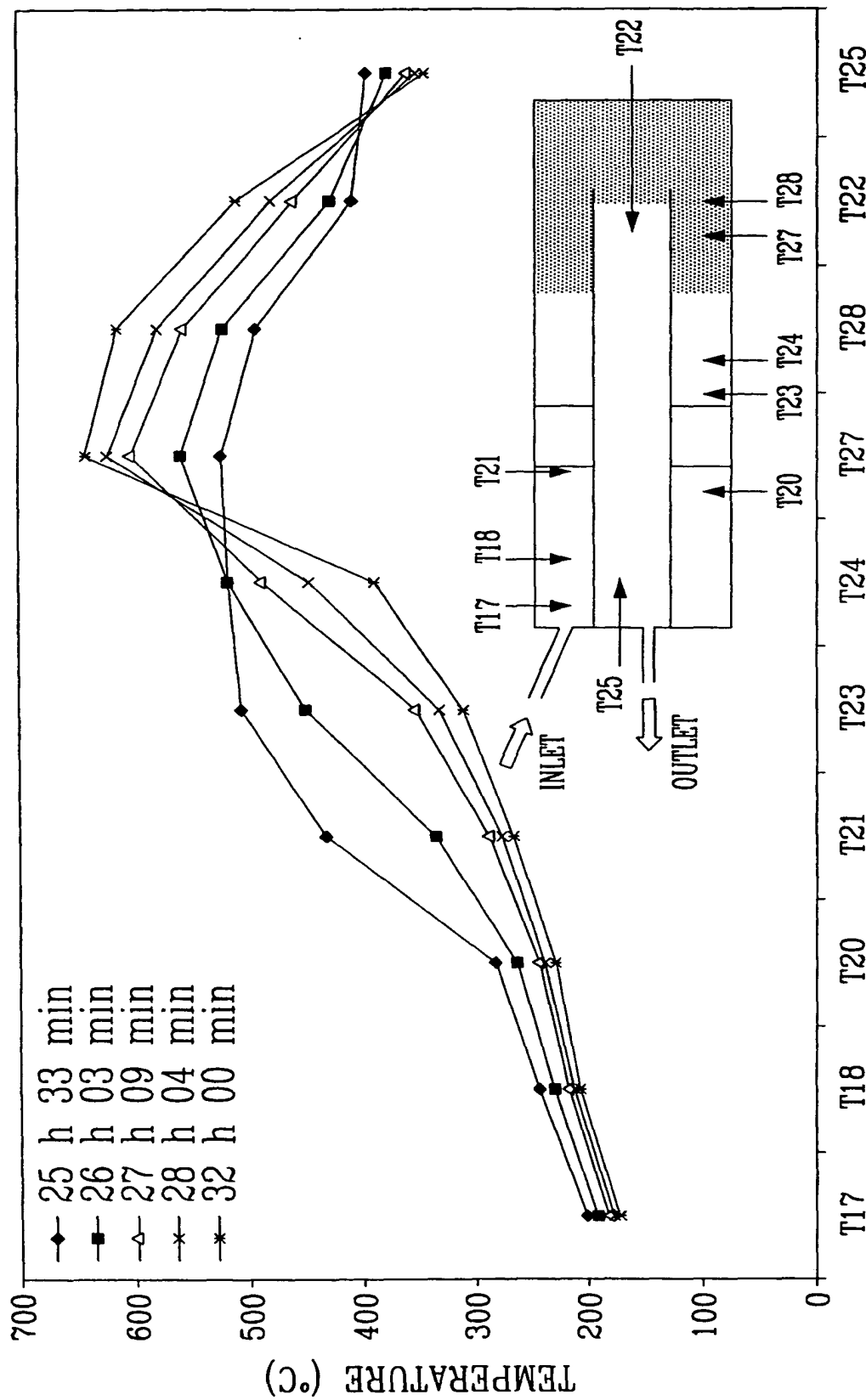


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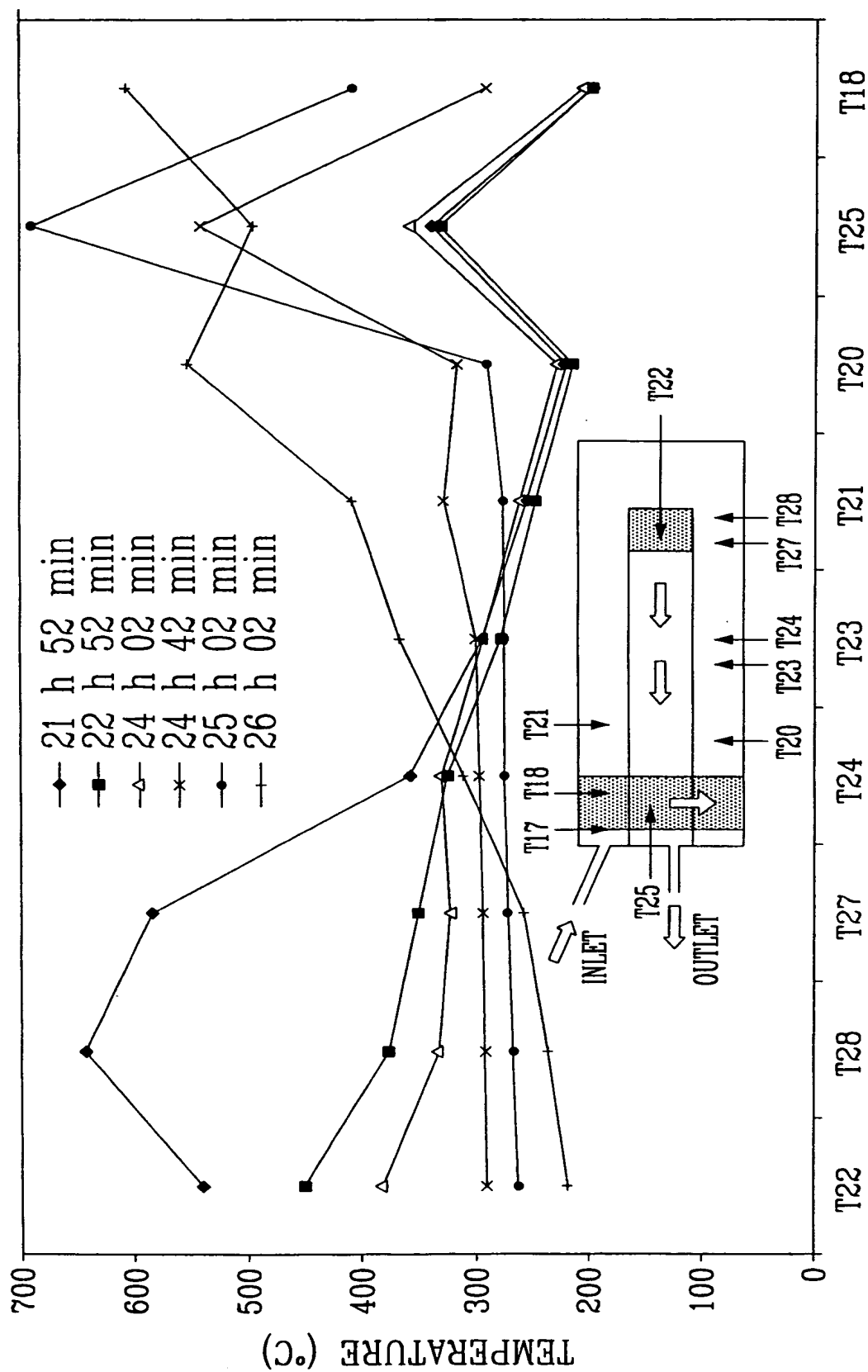


THERMOCOUPLES

FIG. 10

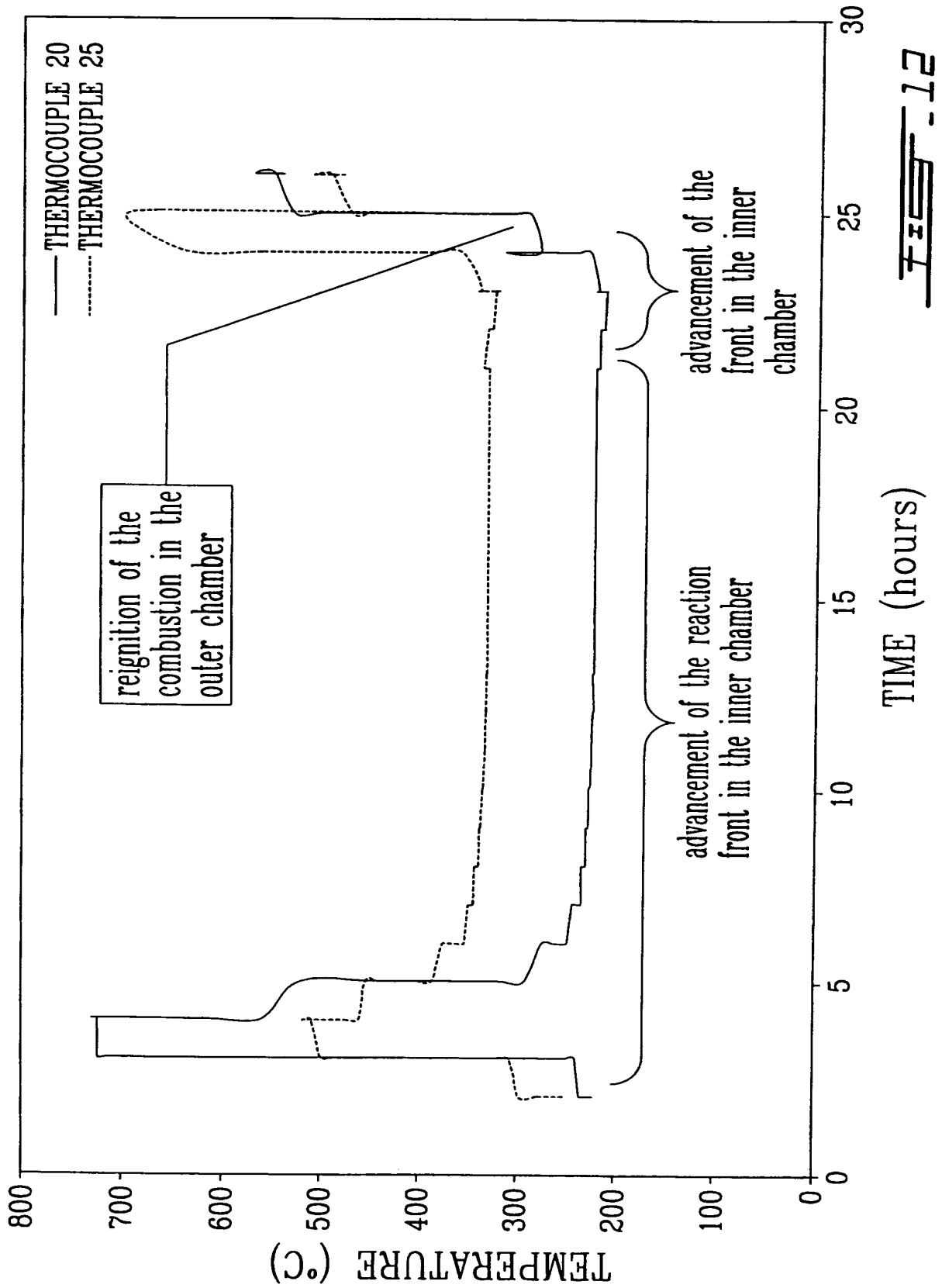


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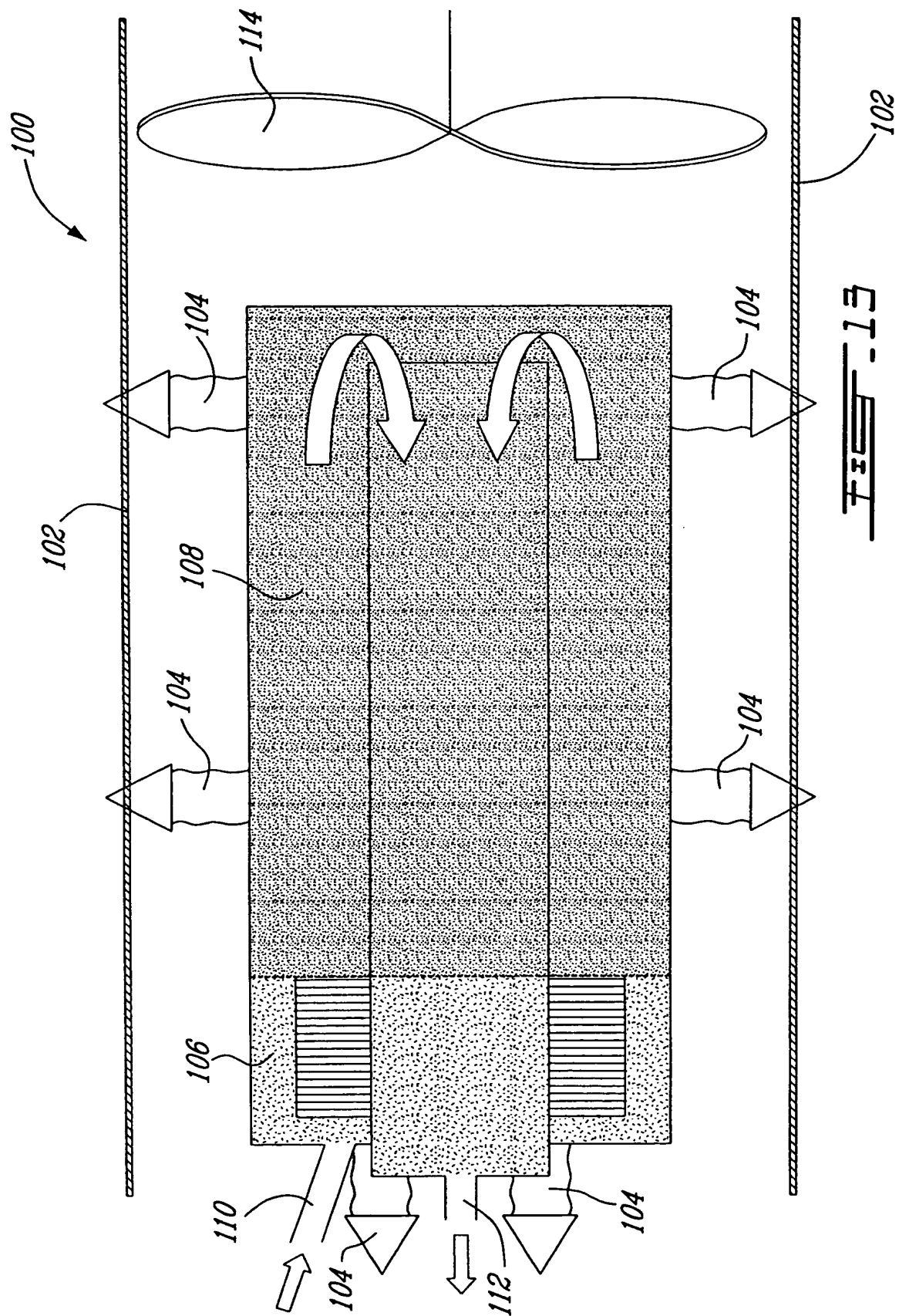
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# INTERNATIONAL SEARCH REPORT

International Application No

PC 00/00713

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 B01J8/04 F23C11/00 F23G7/06

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 B01J F23G F28D F23C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

| Category * | Citation of document, with indication, where appropriate, of the relevant passages  | Relevant to claim No. |
|------------|---|-----------------------|
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☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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Date of the actual completion of the international search

16 October 2000

Date of mailing of the international search report

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# INTERNATIONAL SEARCH REPORT

International Application No

PCT/00/00713

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Information on patent family members

International Application No

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